



## Conference Paper

# Domain Shape Appeared in Stoichiometric Lithium Niobate as a Result of Ion Beam Irradiation

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## Abstract

We have studied the formation of isolated domains induced by ion beam irradiation in the stoichiometric lithium niobate (SLN) single crystals covered by surface dielectric layer. The unusual domain shape was revealed at the irradiated polar surface at the doses above 20 pC. The nested domain shape with hexagonal outer part and circle inner one has been distinguished. The domains visualization in the bulk showed the hexagonal domain shape in the depth. The obtained effect was attributed to backswitching under the action of electric field produced by space charge dipped to LN plate at the doses above 20 pC due to essential ion beam sputtering effect.

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## 1. Introduction

The elaboration of the ways to modify the properties of nonlinear-optical crystals by creation of ferroelectric periodic micro- and nanodomain structures is developed rapidly at present time [1-4]. The periodically poled domain structures (PPDS) can be used for the creation of new types of electro-optical and nonlinear-optical devices. The lithium niobate (LiNbO<sub>3</sub>, LN) crystals are the most attractive materials for these applications due to its unique physical properties [5]. The high quality LN crystals with stoichiometric composition can be considered as model ferroelectrics due to low concentration of point defects [6]. Moreover, they are very promising materials for optical applications due to enhanced photorefractive damage threshold [7].

The traditional periodical poling is based on applying an external electric field by means of electrodes. This method has some disadvantages such as the domain broadening outside of the area covered by electrodes, domain merging and spontaneous backswitching [8]. These problems stimulate the development of the poling methods using an electron (e-beam) [9-14] and an ion beams (i-beam) [18-20].

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It was shown recently that the e-beam switching is very promising and allows to create of PPDS with high quality and homogeneity but has the difficulties in submicron domain engineering [9-14]. The irradiation by focused i-beam leads to small interaction volume and allows to reach the submicron domain size and structure period [14-17].

Our study is devoted to the development and improvement of non-contact method of the domain structure formation with proper geometric parameters by focused ion beam. We have investigated the formation of isolated domains as a result of irradiation by focused ion beam of the stoichiometric lithium niobate (SLN) single crystals covered by surface photoresist layer.

## 2. Materials and experiment

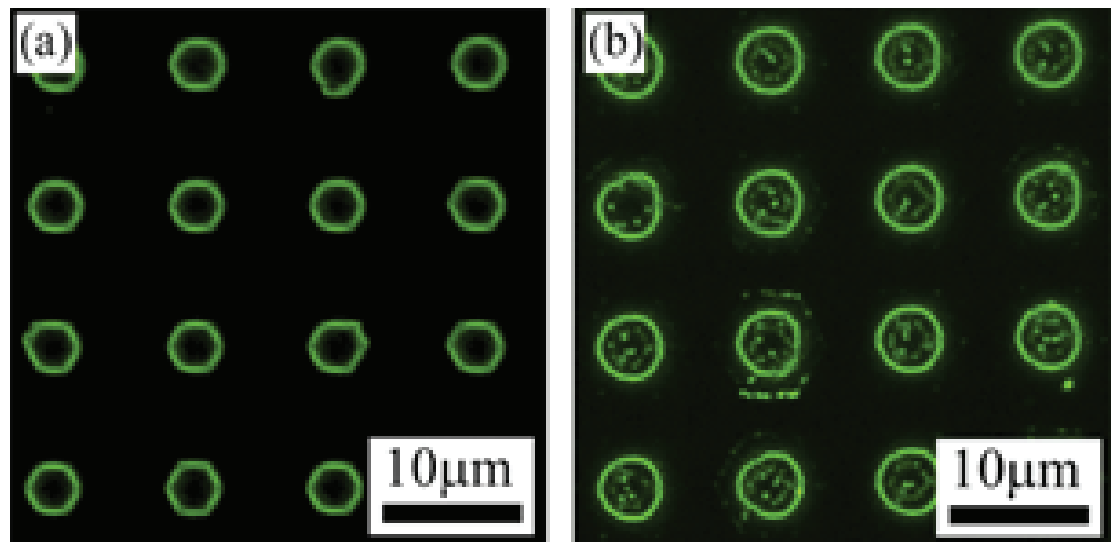
The studied samples represented Z-cut optical grade single crystalline 0.5-mm-thick wafers of SLN (SAES Getters, Italy). The  $Z^+$  polar surface before irradiation was covered by 500-nm-thick layer of negative photoresist deposited by spin coating. The solid 100-nm-thick Cu electrode was sputtered on  $Z^-$  surface and grounded during irradiation.

The  $Z^+$  polar surface was irradiated by ion beam using dual-beam workstation (equipped with electron and ion beam) Auriga Crossbeam (Carl Zeiss NTS, Germany) with liquid-metal ion source of  $Ga^+$  ions using dot irradiation regime. The beam movement and irradiation parameter were controlled by ion-beam lithography system Elphy Multibeam (Raith GmbH, Germany). The samples were irradiated by different doses at fixed accelerating voltage of 30 kV and beam current ( $I$ ) of 1 nA. The dose was defined as  $D = I \times t$ , where  $t$  is irradiation time.

The created isolated domains after chemical removing of the resist and electrodes were revealed by selective chemical etching in pure HF [23] and visualized by several microscopic methods: (1) optical microscopy (OM), (2) piezoresponse force microscopy (PFM) [24], and (3) Cherenkov second harmonic generation microscopy (CSHG) [25-26].

## 3. Results and discussion

The samples were irradiated by beam size about 400 nm using patterns of square arrays of  $10 \times 10$  dots with 10  $\mu\text{m}$  period and dose ranged from 10 to 100 pC. The irradiation results in formation of through isolated domains (Fig. 1). The hexagonal domains appeared at the low doses and the circular ones at the high doses. The domains appeared as sub-micron domains at the bottom polar surface.

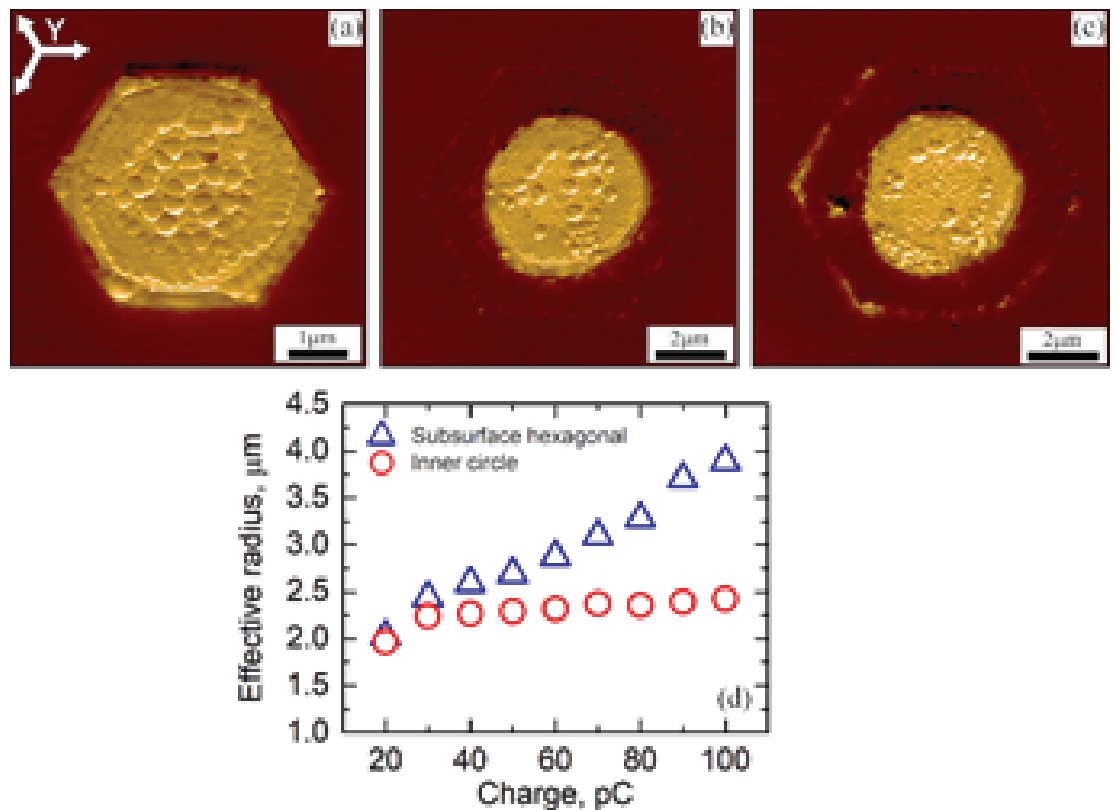


**Figure 1:** Optical images of the isolated domains after ion beam irradiation at  $Z^+$  polar surface with dose of (a) 20 pC, (b) 100 pC. The domains were revealed by etching.

The domain shape was studied with high resolution by PFM. The PFM images showed a hexagonal domain border that surrounds the circular domain at all doses above 20 pC (Fig. 2(a) – 2(c)). Due to integral signal of PFM the signal depth can be down to  $1.7 \mu\text{m}$  [27]. The effective domain size was measured as  $R = (A/\pi)^{1/2}$ , where  $A$  is the domain area. The obtained dose dependences are presented in Figure 2(d). It was shown that the size of inner circular part increased insignificantly (about 15%) while the hexagon radius demonstrated significant growth in the studied dose range.

The shape of domain produced at the high dose was performed in the bulk by CSHG images at the different depths (Fig. 3(a), 3(b)). The CSHG images showed that the domain represents a hexagonal truncated pyramid in the bulk of the crystals while the circular domain at the center of the hexagon located at the polar surface and the hexagonal domain shape restores with depth rapidly (Fig. 3(e)).

The formation of circular domain at the doses above 20 pC can be attributed to backswitching effect [28]. It was shown by us earlier that the space charge of incident ions penetrated through the surface resist layer and dipped in LN plate [21-22] for dose above 20 pC due to i-beam surface sputtering. Thus, different doses lead to different field amplitude and field distribution in subsurface layer. The space charge dipped in LN produce the field oriented in both polar directions: (1) opposite to  $Z^+$  polar direction which led to forward domain growth, and (2) in  $Z^+$  polar direction which led to domain backswitching (Fig. 3(e)). The detail experimental study of the obtained phenomenon and simulations of the electric field distribution will be published by us elsewhere.



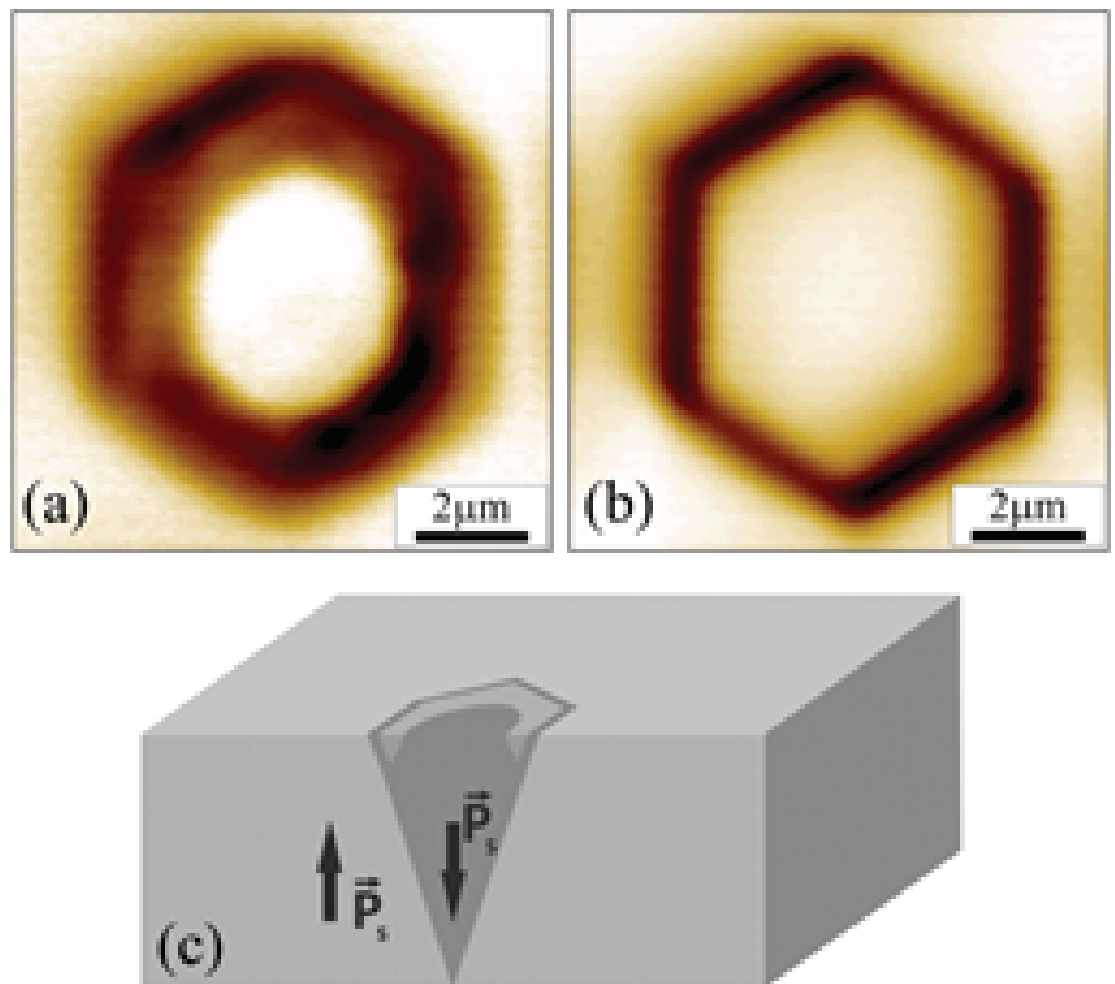
**Figure 2:** PFM images of isolated domains on the  $Z^+$ -polar surface after dot irradiation at doses: (a) 10 pC, (b) 50 pC, (c) 100 pC. (d) The dose dependences of the size of isolated domains (red – inner circular domain size, blue – subsurface hexagonal domain size).

## 4. Conclusions

The formation of isolated domains by focused ion beam in the stoichiometric lithium niobate (SLN) single crystals covered by surface dielectric layer was studied. The visualization of the domains at the surface showed that at the doses above 20 pC the nested domain structure of outer hexagonal part and inner circular part can be distinguished. The visualization of the domain in the bulk showed that the circular domain located at surface only and the hexagonal domain shape rapidly restores with the depth. The effect was attributed to domain backswitching induced by electric field of space charge dipped to LN plate due to ion beam sputtering.

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**Figure 3:** The CSHG images of isolated domains at various depths from  $Z^+$ -polar surface (a) surface, (b) 2 μm, after dot irradiation with dose 100 pC. (c) The scheme of isolated domain in the crystal bulk.

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